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Near- and far field spectroscopy of semi-continuous gold films with optically induced anisotropy

Christian Frydendahl[1,2], Taavi Repän[1], Mathias Geisler[1,2], Sergey M. Novikov[3], Jonas Beermann[3], Andrei Lavrinenko[1], Sergey I. Bozhevolnyi[3], N. Asger Mortensen[2,3] and Nicolas Stenger[2,3]

[1]Department of Photonics Engineering, Technical University of Denmark, DK-2800 Kongens Lyngby, Denmark
[2]Center for Nanostructured Graphene, Technical University of Denmark, DK-2800 Kongens Lyngby, Denmark
[3]Centre for Nano Optics, University of Southern Denmark, Campusvej 55, DK-5230 Odense M, Denmark

E-mail address: chrfr@fotonik.dtu.dk

Abstract: Using electron energy loss spectroscopy (EELS), optical spectroscopy, and finite element method simulations, we investigate the distributions of plasmonic modes present in fractal semi-continuous gold films, as well as studying anisotropy introduced into these films from photothermal melting with a Ti:sapphire laser.

We have used EELS and optical spectroscopy to study the distributions of plasmonic modes present in thin semi-continuous gold films of nominal thicknesses of 5, 6, and 7 nm, as well as the effect of photothermally induced damage from a fs-pulsed Ti:sapphire laser on the distribution of these modes. We have previously observed plasmonic field enhancement, white light generation, and the ability to induce strongly anisotropic damage in these types of samples [1,2].

After illuminating an area of the films with linearly polarized laser pulses above a critical power, we see features in the transmission spectra. These features have a polarization dependence, which is correlated with the polarization of the laser used to induce the photothermal damage, see Fig. 1. Using finite element method simulations of film morphologies recovered from scanning transmission electron microscope (STEM) images, we study the polarization dependence of the field distributions of plasmon modes, and compare this with the recorded EELS near-field optical data.

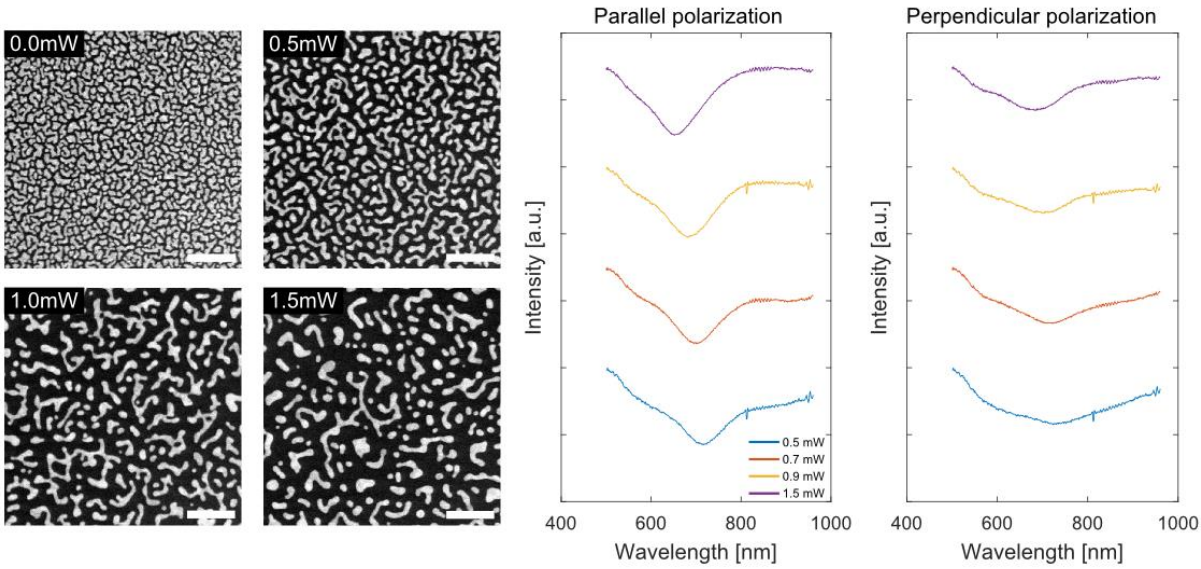

Fig. 1. Left: STEM images of pristine and different laser powers of damage induced in a 5 nm gold film, scale bars are 200 nm. The polarization of the laser used for the damage was orientated along the east-west direction in the images. Right: Bright field transmission spectra from regions of different powers of optical damage. A polarizer has been inserted between the light source and sample. Aligning the polarizer parallel to the polarization of the laser used to induce the damage, shows a peak decrease in transmission. For the perpendicular polarization this peak is severely reduced, showing that large scale anisotropy has been introduced into the sample from the optical damage.

[1] S. Novikov, J. Beermann, C. Frydendahl, N. Stenger, V. Coello, N. Mortensen, and S. Bozhevolnyi, "Enhancement of two-photon photoluminescence and SERS for low-coverage gold films," Opt. Express 24, 16743-16751 (2016)
[2] S. Novikov, C. Frydendahl, J. Beermann, V. Zenin, N. Stenger, V. Coello, N. Mortensen, and S. Bozhevolnyi, arXiv:1701.01839, (2017)